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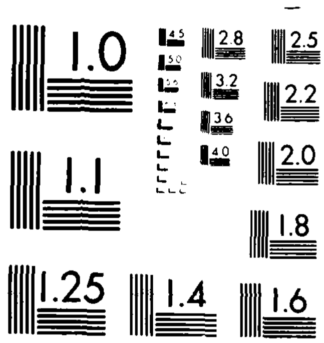

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EFFECT OF NICKEL ADDITION ON THE THERMAL, MAGNETIC AND MAGNETOMECHANICAL PROPERTIES OF $Fe_{80}B_{15}Si_5$

BY L. T. KABACOFF M. WUN-FOGLE

RESEARCH AND TECHNOLOGY DEPARTMENT

2 JULY 1984

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Fe-based metallic glasses with about 20 a/o metalloid (B, Si, and sometimes C) possess very high magnetomechanical coupling factors when properly field annealed. However, the corrosion resistance is very poor, leading to rust formation if left in air for a few weeks or months. Additions of Ni promote improved resistance to corrosion, but would be expected to degrade the magnetostriction somewhat. This report evaluates the effect of Ni additions on some thermal, magnetic and magnetomechanical properties of

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a typical high coupling metallic glass. The Curie temperatures (T_c), magnetomechanical coupling factors (K_{33}), anisotropy fields (H_A), and crystallization temperatures (T_x) of the series of metallic glasses, $Fe_{80-x}Ni_xB_{15}Si_5$ ($0 < x < 40$), were measured. The values of T_c were determined from the specific heat anomaly using a differential scanning calorimeter. K_{33} was determined by a resonance technique. H_A was estimated from the bias field which yields a minimum in resonance frequency (i.e., minimum Young's modulus). T_x, defined by the onset of crystallization, was also measured by DSC, at a heating rate of 20°C/min.

T_c versus a/o Ni exhibits a maximum at 20 a/o Ni. The value of T_c is 389, 438, and 366°C for 0, 20, and 40 a/o, respectively. The value of K_{33}, on the other hand, falls nearly linearly with increasing Ni content, changing by 0.006 for each additional atomic percent Ni. The anisotropy field also goes through a maximum, with values of 1.0, 1.5, 2.0, and 1.5 Oe for 0, 10, 20, and 40 a/o, respectively. Finally, the crystallization temperature falls linearly with increasing Ni content, from 507 to 454°C. The effect of Ni addition on K_{33} and H_A can be related to the change of the magnetostriction and magnetic moment with increasing Ni content.

Supplied keywords

FOREWORD

This work was performed under the Independent Research Program of the Naval Surface Weapons Center (NSWC), and is part of an ongoing program to develop amorphous metal magnetostrictive transducers with very high magnetomechanical coupling factors. In this report, the question of the corrosion resistance of these materials, and, in particular, how to improve corrosion resistance in air without sacrificing transducer properties, is addressed. The results presented here should be of value to those interested in designing sensors which utilize these new materials. The results presented here detail the effect of nickel additions on the thermal and magnetic properties of a typical amorphous alloy with outstanding transducer properties. The trade-offs between transducer and corrosion properties are discussed as well as opportunities for further improvement.

Approved by:

J. R. Dixon

JACK R. DIXON, Head
Materials Division

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ILLUSTRATIONS

CHAPTER 1

INTRODUCTION

In recent years there has been a great deal of interest in ferromagnetic metallic glasses due to their unique magnetic and magnetomechanical properties. Of these, the property of greatest interest to the Navy is the fact that, when properly annealed, these materials possess the highest value of magneto-mechanical coupling, K_{33}, known--greater than 0.9[1] (as compared with 0.3 for nickel and 0.75 for Terfenol[2]). K_{33}^2 is equal to the efficiency with which magnetic and magnetomechanical energy are interconverted. This high coupling factor, combined with excellent soft magnetic properties, makes ferromagnetic metallic glasses a very promising material for use as transducer elements in a wide variety of sensors.

One problem with these materials which has become apparent is that they are prone to corrode with rust visible in a few weeks or months if samples are not kept in a dry atmosphere. Thus, while it is possible to design a system in such a way that the transducer is protected from moisture, it would be convenient to find compositions which are more corrosion resistant. It is highly unlikely that corrosion resistance to sea water is attainable without sacrificing most or all of the advantages of metallic glasses. However, a high coupling material which will not corrode in humid air is quite possible.

The high coupling metallic glasses have compositions of the type Fe_xM_{100-x} where x is near 80 a/o and M consists usually of B and Si and, sometimes, C (P has not been fully investigated, but would likely segregate during annealing). The three elements which could most likely be added to enhance corrosion resistance are Cr, Co, and Ni. Cr has the disadvantage of coupling to the Fe antiferromagnetically. This leads to a sharp decrease in the average magnetic moment and Curie temperature and, therefore, K_{33}. However, in some applications not requiring very high coupling, Cr is used. The disadvantage of Co is that it greatly increases the uniaxial anisotropy energy of the material, as well as the local variation in the direction of the magnetic easy axis. This leaves Ni, which has a number of properties, which indicate that Ni, high K_{33} materials might exist. For one thing, Ni does not necessarily reduce the Curie temperature of an Fe-based metallic glass. Also, though Ni does not tend to lower the saturation magnetization somewhat, it does so much less precipitously than Cr. Finally, Fe-based glasses with significant amounts of Ni are good glass formers. For all of these reasons, an initial study has been made of the effect of Ni on the Curie temperature, magnetomechanical coupling, anisotropy field and crystallization temperature of a typical high coupling Fe-based metallic glass, $Fe_{80-x}Ni_xB_{15}Si_5$.

CHAPTER 2

EXPERIMENT

ALLOY PREPARATION

Master ingots of $Fe_{80}B_{15}Si_5$ and $Fe_{40}Ni_{40}B_{15}Si_5$ were arc melted under argon atmosphere from starting materials of m3N purity or better. Appropriate portions of the two ingots were arc melted together to form intermediate compositions with 10, 15, and 20 a/o Ni. These were then meltspun in air to form amorphous metallic ribbons approximately 0.5 mm wide and 20 μm thick. A schematic of the meltspinner is shown in Figure 1. Samples were checked for the presence of crystallinity by X-ray diffraction, using Mo Kα radiation. All compositions formed glasses easily.

DIFFERENTIAL SCANNING CALORIMETRY (DSC)

Small pieces (between 4 and 15 mg) were cut up, weighed and encapsulated in aluminum sample pans. DSC curves (which measure heat flow into or out of the sample as a function of temperature) were then obtained at a heating rate of 20°C/min. A typical curve is illustrated in Figure 2.

The Curie temperature, T_c, was measured by noting the minimum in the endothermic specific heat anomaly associated with the Curie transition.[3] This is a true zero field measurement. However, since T_c depends on the state of structural relaxation and, therefore, thermal history, the value obtained is somewhat dependent on heating rate. This effect was neglected in the present work since it is small compared to the effect of Ni addition to the composition.

The crystallization temperature, T_x, was determined by the "onset" method, in which the steepest slope of the crystallization exotherm is extrapolated to the baseline. T_x is dependent on the heating rate, and comparisons should only be made between values of T_x obtained at a similiar heating rate.

MAGNETOMECHANICAL PROPERTIES

Metallic glass ribbons were cut into 3-inch lengths and annealed at various temperatures in a saturating magnetic field oriented in the plane of the ribbon transverse to the length. Samples were then characterized magnetomechanically using the set-up illustrated in Figure 3.[2] The ribbon is placed in a pick-up coil and subjected to both a DC bias field, H_B, and a small AC field. For each value of H_B, the frequency of the AC field is swept, and the voltage in

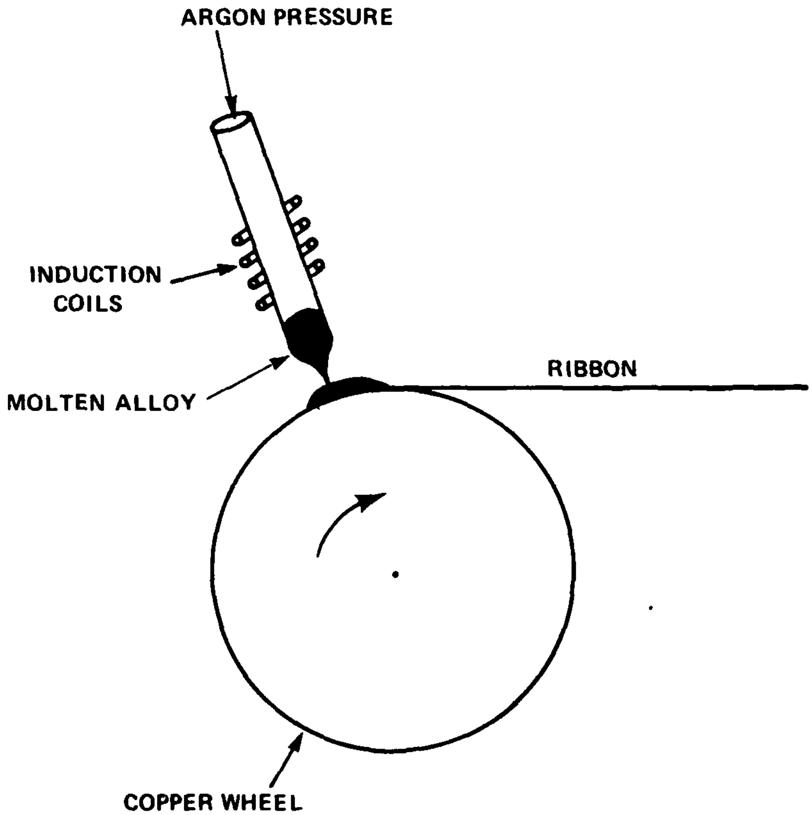

FIGURE 1. APPARATUS FOR MELTSPINNING AMORPHOUS METAL RIBBONS

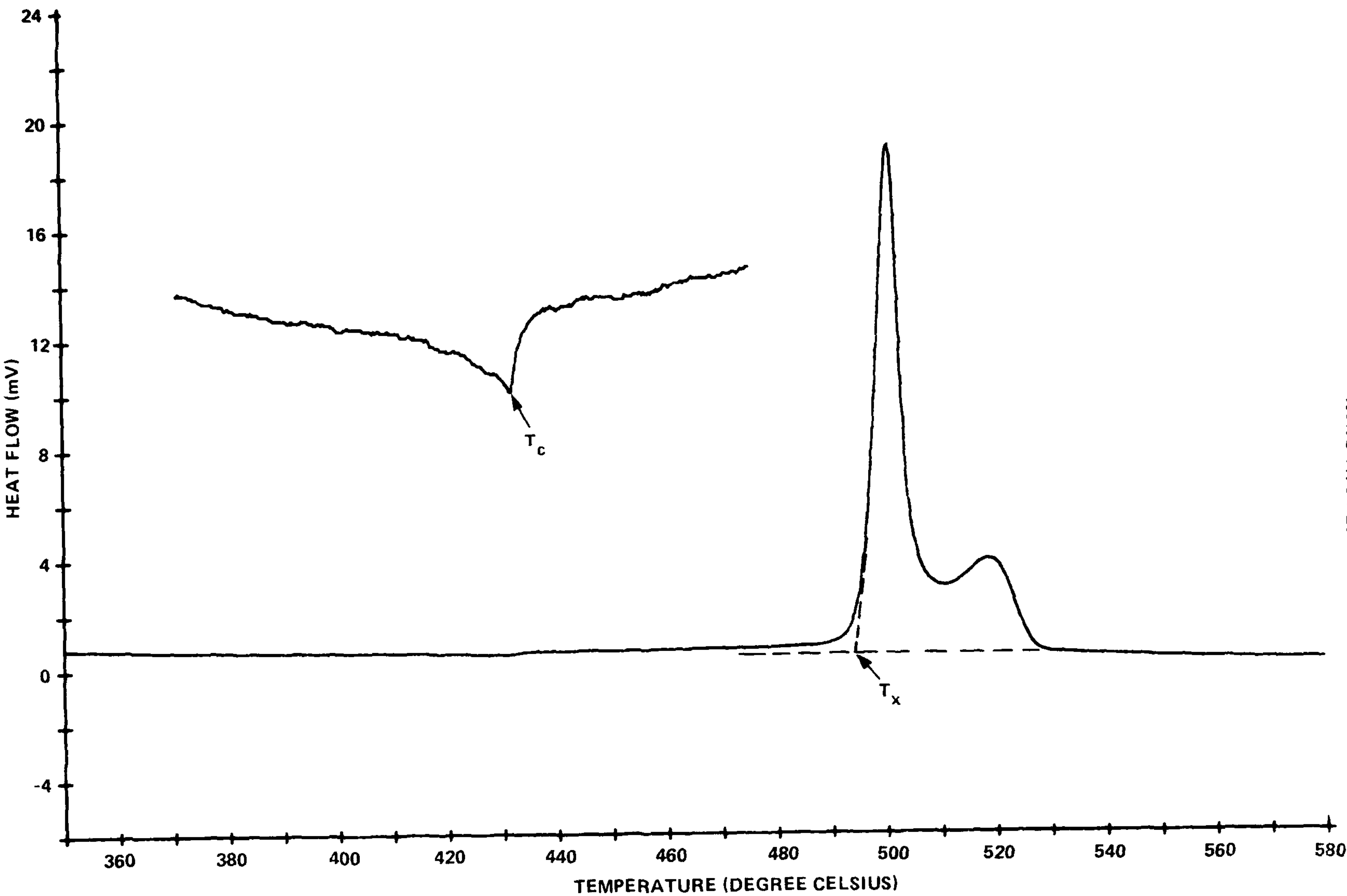

FIGURE 2. TYPICAL DSC CURVE FOR A FERROMAGNETIC METALLIC GLASS

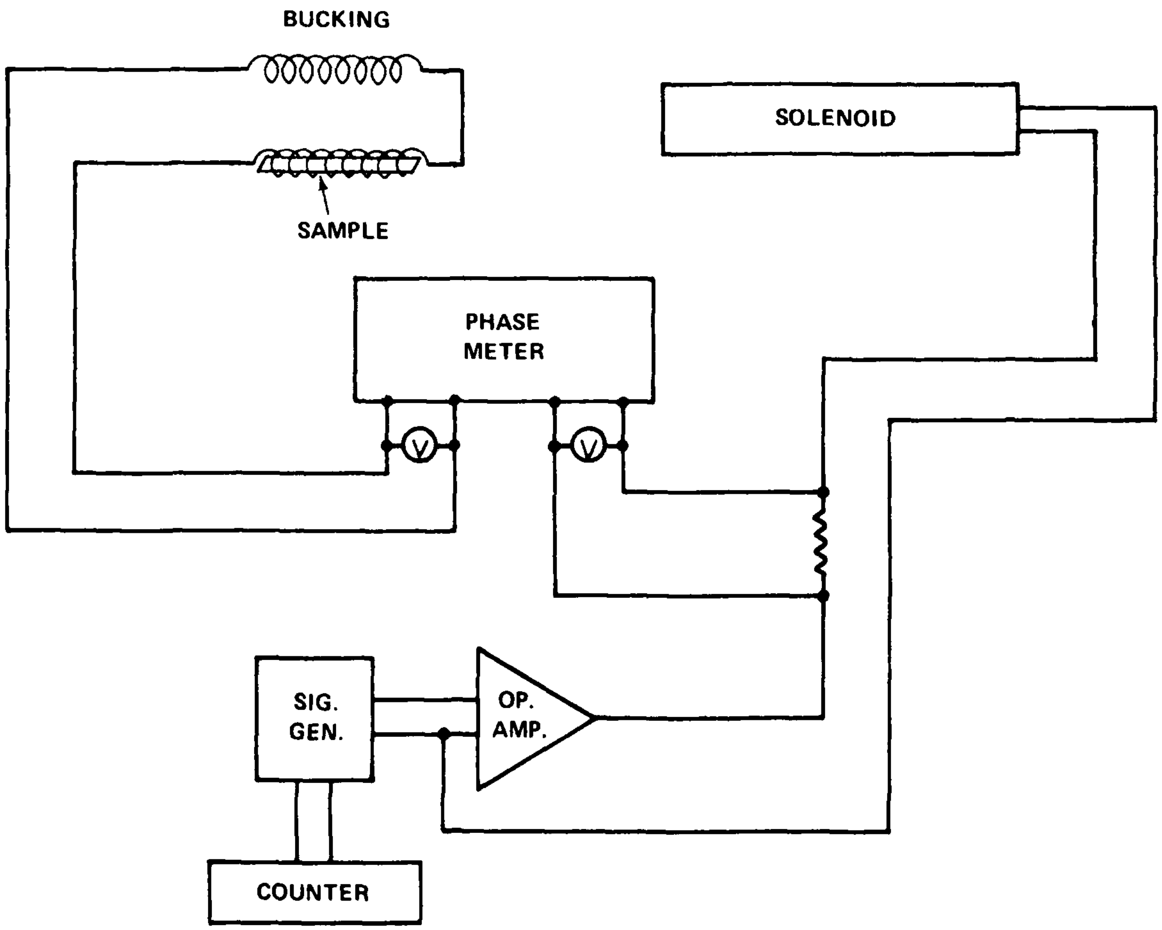

FIGURE 3. APPARATUS FOR MEASURING THE MAGNETOMECHANICAL PROPERTIES OF MAGNETOSTRICTIVE MATERIALS

the pick-up coil measured. The maximum and minimum in this voltage define the resonance and antiresonance frequencies, which are plotted versus H_B. The value of H_B which gives the minimum resonance frequency is the anisotropy field. The magnetomechanical coupling factor, K_{33}, is calculated from the equation,[1]

$$K_{33}^{\ 2} = \frac{\pi^2}{8}\left(1 - \frac{f_r^{\ 2}}{f_a^{\ 2}}\right) \qquad (1)$$

where f_r and f_a are the resonance and antiresonance frequencies respectively. A bucking coil is placed in series with the pick-up coil to cancel the flux directly linking the drive and pick-up coils.

CHAPTER 3

RESULTS AND DISCUSSION

The effect of Ni addition on the Curie temperature, T_c, is illustrated in igure 4. The value of T_c initially increases, from 389°C to a maximum of 37°C at 20 a/o Ni. T_c then decreases monotonically to a value of 366°C at 40 /o Ni. It is interesting to note that the magnetic moment per transition metal tom falls linearly over this region, from 2 μ_B to about 1.2 μ_B. A similiar ncrease is observed when Ge is substituted for Si.[4] In that case the increase s attributed to an increase in the Fe-Fe interatomic distance. In the present ase, it is likely that size effects and moment dilution compete, yielding a aximum in T_c. Figure 4 also shows the crystallization temperature, T_x, as a unction of Ni content. The value of T_x falls approximately linear with ncreasing a/o Ni, from 507 to 454°C. None of the samples exhibited a measurable lass transition except for the 40 a/o Ni sample, which showed a very pronounced lass transition. This indicates that the activation energy of crystallization, hich correlates with the appearance of glass transition, is increasing with ncreasing a/o Ni. Thus, the decrease in T_x is due to a change in the requency factor.

The anisotropy field, which is illustrated in Figure 5, also has a maximum t approximately 20 a/o Ni. The value of H_a is related to the saturation agnetization and uniaxial anisotropy energy by the expression:

$$H_a = 2K_u/M_D \quad . \tag{2}$$

he subscript, D, refers to a domain average. As noted above, M_D decreases pproximately linearly with Ni content. Thus, K_u has a maximum which, like he Curie temperature, results from competing effects. Initially, K_u ncreases because the addition of Ni into what were previously Fe sites is very asily accomplished in an anisotropic manner. In other words, if the irectional distribution of Ni-Fe pairs are skewed, a magnetic anisotropy will esult which would not exist if Ni was absent. Such skewness is, of course, roduced by the magnetic anneal. However, as the Ni concentration increases, he dilution of the magnetization by the Ni begins to reduce the value of K_u. hus, one obtains a maximum.

Figure 6 shows the effect of Ni concentration on the maximum magneto-echanical coupling factor, K_{max}. K_{max} is defined as the highest value of $_{33}$ which can be obtained for a given composition by optimizing annealing emperature and bias field. As may be seen in the figure, K_{max} decreases pproximately linearly with increasing Ni content. This linearity is most

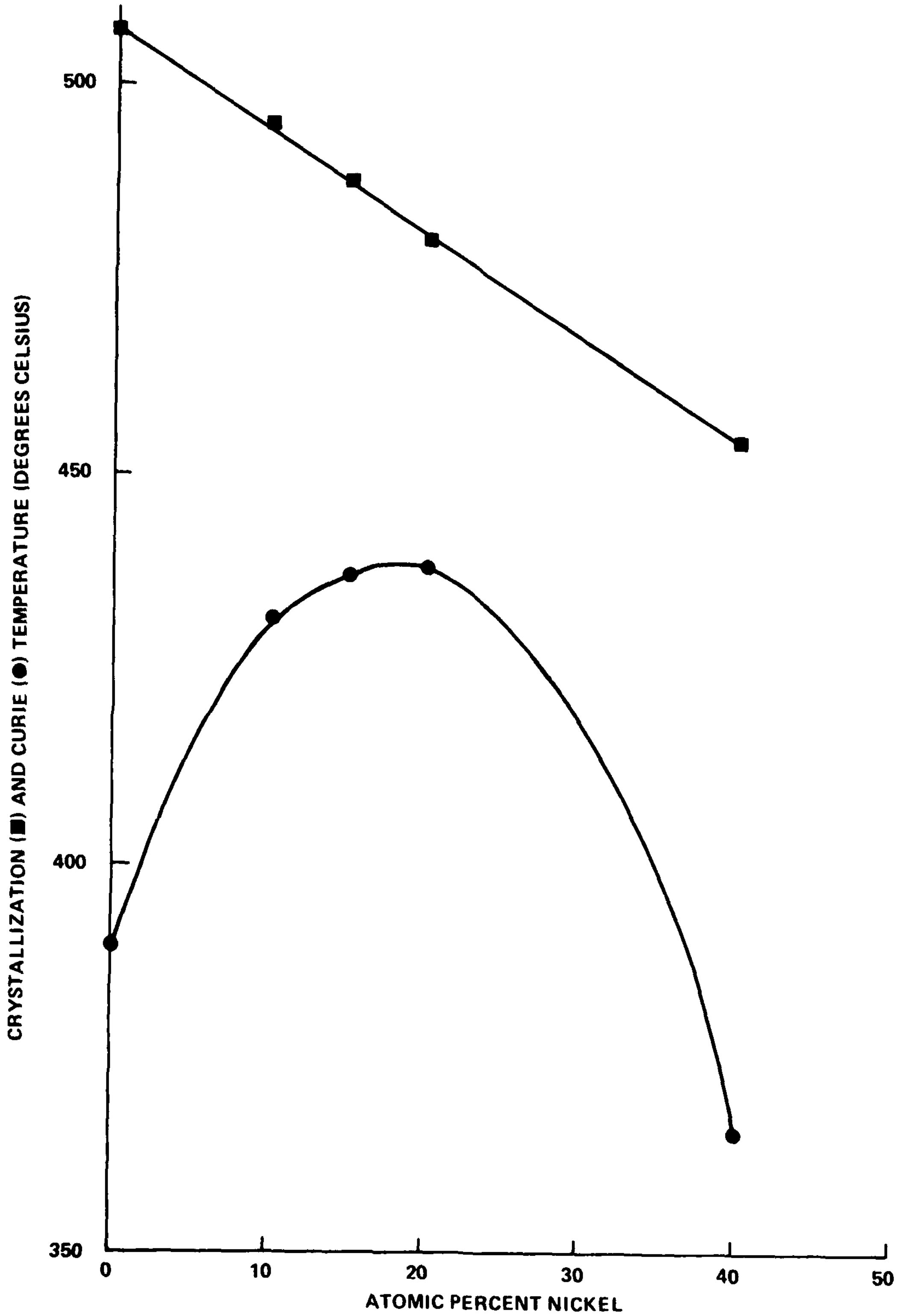

FIGURE 4. CRYSTALLIZATION AND CURIE TEMPERATURES VERSUS ATOMIC PERCENT NICKEL

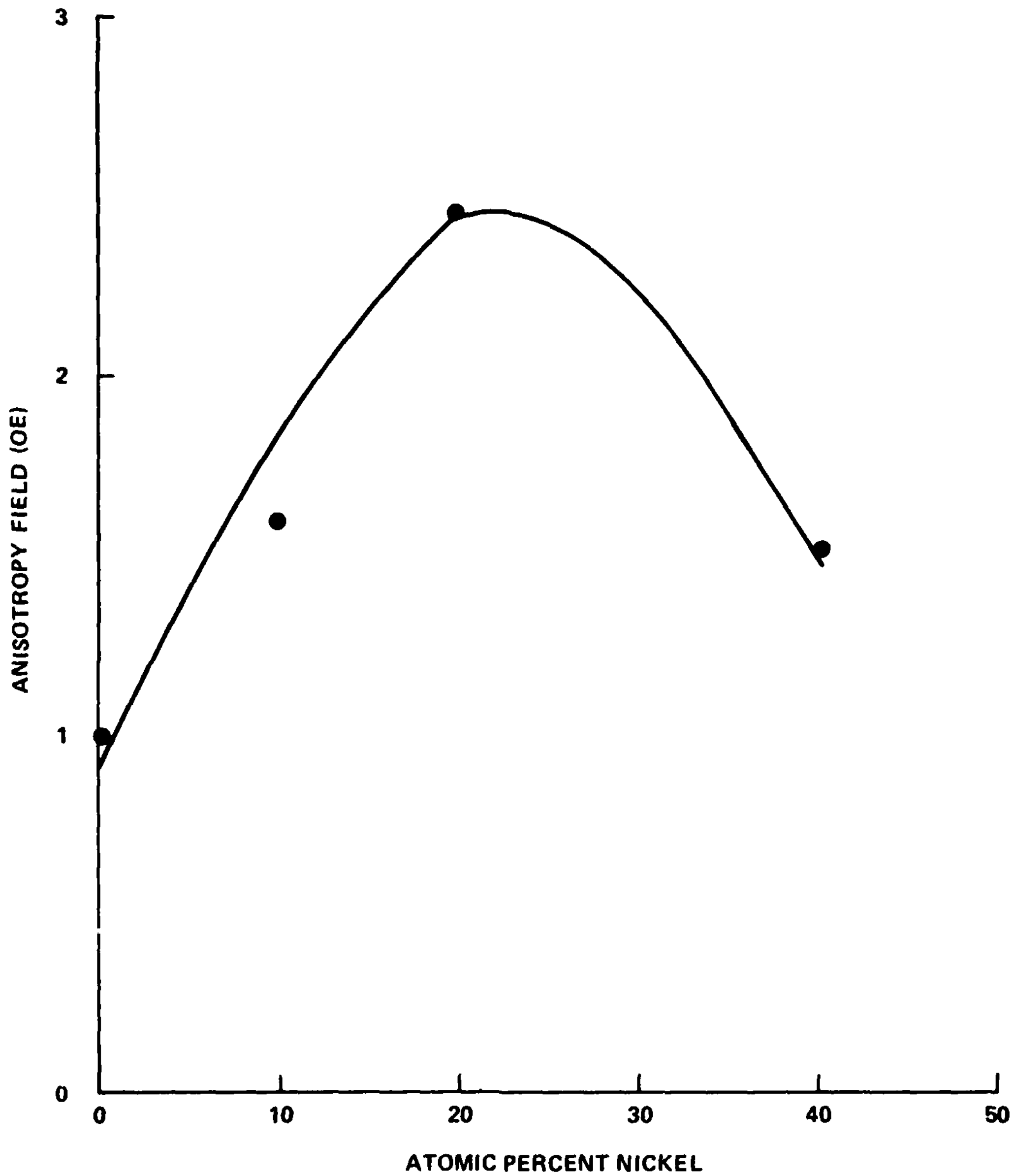

FIGURE 5. ANISOTROPY FIELD VERSUS ATOMIC PERCENT NICKEL

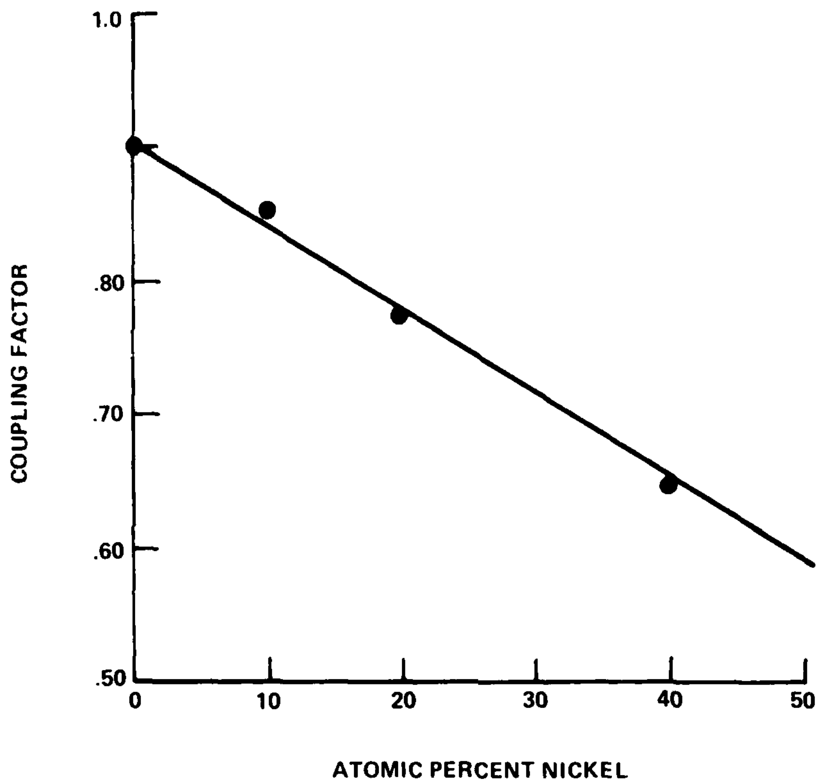

FIGURE 6. MAXIMUM MAGNETOMECHANICAL COUPLING FACTOR VERSUS ATOMIC PERCENT NICKEL

likely coincidental since K_{33} by itself is not a physically meaningful parameter. A more meaningful figure of merit is given by $K^2/(1 - K^2)$, which is related to the fractional change in the Young's modulus as a function of the applied bias field. The slope of the K_{max} versus Ni concentration plot is 0.006 per a/o Ni.

To a reasonable approximation, the coupling factor is given by:

$$K_{33}^{2}\Big/\left(1 - K_{33}^{2}\right) = \frac{(3\lambda_s)^2\ Y}{2\ K_u}\left(\frac{H}{H_a}\right)^2 \qquad (3)$$

where λ_s is the saturation magnetostriction, Y is the Young's modulus for a magnetically saturated sample, H is the applied field and K_u is the uniaxial anisotropy. By definition, H/H_a is equal to one for K_{33} equal to K_{max}. The monotonic decrease of K_{max} with increasing a/o Ni parallels the behavior of λ_s with increasing Ni content. There is no evidence of the maximum in K_u affecting the curve of K_{max} versus a/o Ni. Thus, it may be concluded that it is the lowering of the saturation magnetostriction which is responsible for the lowering of K_{max} with increasing Ni addition.

CHAPTER 4

CONCLUSIONS

The results which have been previously described indicate that the addition of Ni to high coupling Fe-B-Si metallic glasses will lower the value of the maximum magnetomechanical coupling factor. The decrease is not precipitous, as in the case of Cr. The amount of Ni which could be tolerated depends on the application. Although no quantitative testing of corrosion properties was done, it was noted that as little as 10 a/o Ni was sufficient to prevent rusting of samples left exposed to air for long periods of time.

Further improvements in the coupling factor for glasses containing Ni would result if a means were found to increase the magnetostriction without overly increasing the anisotropy. Some investigation of this possibility has begun with a study of the effect of small Pr additions on Fe-B-Si metallic glasses. However, while the results were encouraging, this was only a small first step. The amount of effort which will be required to complete this effort is sufficiently great that it will be funded only if a specific need is demonstrated. It will be prudent to determine if specific applications will require both maximum coupling factor and corrosion resistance. Once this is demonstrated, funding should be made available to develop the needed material.

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